

Controls on fluxes and export of dissolved organic carbon in grasslands with contrasting soil types

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Abstract Export of dissolved organic carbon (DOC) from grassland ecosystems can be an important C flux which directly affects ecosystem C balance since DOC is leached from the soil to the groundwater. DOC fluxes and their controlling factors were investigated on two grassland sites with similar climatic conditions but different soil types (Vertisol vs. Arenosol) for a 2.5-year period. Parts of both grasslands were disturbed by deep ploughing during afforestation. Contrary to what was expected, ploughing did not increase DOC export but surface soil DOC concentrations decreased by 28% (Vertisol) and 14% (Arenosol). DOC flux from the soil profile was negatively influenced by the clay content of the soil with seven times larger DOC export in the clay-poor Arenosol ($55 \text{ kg C ha}^{-1} \text{ a}^{-1}$) than in the clay-rich Vertisol ($8 \text{ kg C ha}^{-1} \text{ a}^{-1}$). At the Arenosol site, highest DOC concentrations were measured in late summer, whereas in the Vertisol there was a time lag of several months between surface and subsoil DOC with highest subsoil DOC concentrations during winter season. DOC export was not correlated with soil organic carbon stocks. Large differences in ^{14}C concentrations of 22–40 pMC between soil organic carbon and DOC in the subsoil indicated that both C pools are largely decoupled. We conclude

that DOC export at both sites is not controlled by the vegetation but by physicochemical parameters such as the adsorption capacity of soil minerals and the water balance of the ecosystem. Only in the acidic sandy Arenosol DOC export was a significant C flux of about 8% of net ecosystem production.

Keywords Afforestation · Dissolved organic matter · Disturbance · C sequestration · Radiocarbon

Introduction

Dissolved organic carbon (DOC) is an important transitional stage between soil organic carbon (SOC) and carbon dioxide (CO_2) in terrestrial ecosystems. DOC is the main form in which organic carbon is transported downwards into the subsoil where it can be mineralised, stabilised or further leached to the groundwater. DOC export from terrestrial ecosystems can be an important carbon (C) flux in the global C cycle (Hope et al. 1994; Siemens 2003). However, DOC also represents a readily available substrate for microorganisms containing a mineralisable proportion of up to 95% (Don and Kalbitz 2005). Only if organic carbon is dissolved it can be incorporated into microorganisms and may be mineralised. Whether DOC is leached or mineralised depends on soil characteristics (pH, texture, aggregation),

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biological constraints (accessibility for microbial community, composition of the decomposer community) and the contact time between substrate (DOC) and decomposer community. Microbial degradation of DOC is a kinetically limited two-step process, starting outside the organisms with cleaving of complex molecules by extracellular enzymes released by microorganisms. Thereafter, cleaved molecules <600 Da can be incorporated and mineralised by microorganisms as source of energy (White 2000). Not only DOC consumption (mineralisation) but also DOC production is coupled to the microbial activity (Kalbitz et al. 2000). DOC may directly derive from the decomposition process if it is either unpalatable and therefore remained or washed away before complete incorporation and mineralisation by the decomposers. Thus, solid soil organic C can be solubilised by microbial activity. DOC can then be transported down the soil profile with mineral soil serving as a chromatographic column at which DOC is adsorbed replacing older DOC or other anions (Guggenberger and Zech 1993). Thus, DOC in subsoil may largely derive from the subsoil itself and only in preferential flow paths and under storm flush conditions surface soil DOC directly reaches the subsoil (Fröberg et al. 2007; Kaiser and Guggenberger 2005).

Leaching of DOC may account for a significant C flux in terrestrial ecosystems (Siemens 2003). However, most studies on DOC fluxes and dynamics (seasonal variability) were carried out in forest or peatland ecosystems where the organic layer (forest floor) and organic horizons are the major source of DOC (Don and Kalbitz 2005; Hope et al. 1994). In grassland systems organic layers are small or totally missing. Stronger seasonality in grasslands compared to forests lead to higher variability of soil moisture (frequent drying and rewetting) and high amplitude of soil temperature, which both may enhance DOC

production (Kalbitz et al. 2000). Studies on DOC fluxes of grasslands are rare even though grasslands comprise 30% of European ecosystems and store as much organic C as forest soils (Arrouays et al. 2001).

Grassland management often includes regular ploughing to sustain soil fertility and to suppress certain weed species. Conversion of grassland into other land use types is an additional ongoing process in Europe and North America that considerably contributes to agriculture greenhouse gas emissions (Janssens et al. 2003). Also for grassland conversion into forest (afforestation) soils are commonly completely or partly ploughed to prepare the seedbed for planted seedlings (Querejeta et al. 2001; Thuille and Schulze 2006). Ploughing causes considerable disturbance which results in soil C losses of 20–50% (Guo and Gifford 2002; Mann 1986). Soil disruptions such as ploughing influence also DOC production and transport (Hornung et al. 1986). However, quantifications of DOC loss after grassland ploughing are missing. Changing DOC discharge of rivers urges the need to enhance our knowledge on DOC production and leaching also for agricultural systems. Finally, DOC fluxes from different types of land use, including grasslands, have to be incorporated into global C models to understand past and future terrestrial C balances. To understand DOC dynamics of different ecosystems field studies have to be conducted since the complexity of DOC production, consumption and transport in soils cannot be simulated appropriately in laboratory studies.

This paper discusses the following controlling factors for DOC concentrations and fluxes at two grassland sites (1) meteorological factors and their seasonality, (2) soil properties, (3) disturbance by ploughing and (4) water balance and seepage water fluxes.

Table 1 Site characteristics, seepage water fluxes and DOC fluxes of the grasslands in Mehrstedt and Kaltenborn

Site	Coordinates	Elevation (m a.s.l.)	Annual precipitation (mm)	Mean annual temperature (°C)	Soil type FAO	Seepage water (mm a ⁻¹)	DOC flux (kg ha ⁻¹ a ⁻¹)	
							10 cm depth	65/110 cm depth
Mehrstedt	10°39'E, 51°16'N	280	547	8.0	Vertisol	177	19	8
Kaltenborn	10°13'E, 50°47'N	330	566	7.8	Arenosol	290	129	55

Materials and methods

Two grassland sites

The two investigated grassland sites “Mehrstedt” and “Kaltenborn” are situated within a distance of 60 km in Thuringia/Germany and are part of the BIOTREE experiment (Scherer-Lorenzen et al. 2007) (Table 1). Both sites had similar above-ground biomass production and were converted into forest by afforestation during the first year of measurement. The planted trees did not have any impact on the ecosystem due to their small size and cover compared to grasses and herbs. Rows of about 30 cm width and 2 m distance from each other were ploughed to 60 cm depth with deep spate moulder to prepare for tree planting. The stripes between planting rows remained undisturbed and allowed a comparison between ploughed and non-ploughed treatments in close vicinity, thus, without the confounding factor of soil heterogeneity.

Both sites have similar meteorological features (Table 1) and land use history (Don et al. 2007). At Mehrstedt, clay-rich Vertisols developed on Triassic limestone covered by loess layers of different thickness (soil characteristics in Table 2). At Kaltenborn

soils are classified as Arenosols with a non-aggregated, single-grain soil structure (Table 3).

DOC sampling and analysis

To investigate grassland DOC fluxes, suction plates were installed at 10 cm of depth (Mehrstedt $n = 11$, Kaltenborn $n = 8$) and at the border between lower B horizons and the bedrock (C horizon) ($n = 16$). This depth (65 and 110 cm depth at both soil types) was defined as the outer border of the ecosystem and is named B–C border throughout this manuscript. DOC fluxes at this border were considered as C losses from the system. Suction plates were distributed at both sites in 4 sampling plots within a distance of <350 m. At each site two plots had a soil thickness of 65 cm (B–C border) and two plots had 110 cm soil thickness. A paired sampling design was used with a corresponding suction plate in the disturbed ploughing row (Mehrstedt $n = 13$, Kaltenborn $n = 12$) and the non disturbed grassland stripes in between (Mehrstedt $n = 14$, Kaltenborn $n = 12$). Additional plates were installed in 30 cm depth at the Mehrstedt site but not used for DOC flux calculations.

Table 2 Soil parameters at the Mehrstedt site (adapted from Don et al. 2007)

Depth (cm)	Horizon	Sand (%)	Silt (%)	Clay (%)	Bulk density	pH (KCl)	C _{org} (g kg ⁻¹)	C/N	CEC (cmolc kg ⁻¹)	Fe _d (g kg ⁻¹)	Fe _o (g kg ⁻¹)	Al _o (g kg ⁻¹)
0–9	Ah	3	62	35	1.23	7.0	25.1	10.2	28	14	2.2	1.4
9–17	Ap	3	63	34	1.48	6.8	13.9	9.3	27	16	2.4	1.4
17–36	Ap-Bi	3	61	36	1.58	6.9	9.9	9.0	31	16	2.4	1.5
36–46	Bdi1	5	43	52	1.70	7.0	4.4	7.0	46	19	1.9	1.3
46–61	Bdgi1	2	28	70	1.90	7.2	3.3	ND	50	14	2.6	1.0
61–100	Bdgi2	2	31	67	1.96	7.4	2.0	ND	45	12	3.5	0.7

Table 3 Soil parameters at the Kaltenborn site (adapted from Don et al. 2007)

Depth (cm)	Horizon	Sand (%)	Silt (%)	Clay (%)	Bulk density	pH (KCl)	C _{org} (g kg ⁻¹)	C/N	CEC (cmolc kg ⁻¹)	Fe _d (g kg ⁻¹)	Fe _o (g kg ⁻¹)	Al _o (g kg ⁻¹)
0–8	Ah	80	11	9	0.97	5.0	21.4	11.2	4	3	0.5	0.5
8–27	Ap	81	10	9	1.39	4.5	7.1	10.4	6	3	0.8	0.6
27–38	Bw1	78	14	8	1.61	4.7	2.3	9.4	4	2	1.2	0.7
38–75	Bw2	77	16	7	1.72	4.3	2.0	9.1	2	2	1.3	0.8
75–110	2Bw	79	14	7	1.78	4.2	0.1	7.2	3	2	0.9	0.7

Borosilicate glass suction plates were used (Eco-tech, Germany) with 1.0–1.6 μm pore diameter and 80 mm plate diameter. Suction plates were constructed without any glue to prevent any contamination of the soil solution samples with organic substances (Siemens and Kaupenjohann 2003). Compared to the commonly used suction cups, suction plates sample preferential flow paths more reliable because of greater area (Siemens et al. 2003) and glass suction plate avoid DOC sorption by the samplers. Suction plates were installed with minimum disturbances to the overlying horizons by excavating soil pits from which >40 cm long horizontal holes (about 10 \times 10 cm) were caved into the soil pit walls. Suction plates were placed onto the upper wall of these holes. To insure optimal contact between suction plate and soil, a suspension of soil material from the appropriate horizon was used to fix the plates. Samples were collected fortnightly in 11 glass bottles with initial 0.4 bar low-pressure applied manually to the bottle-suction plate system to balance the matrix potential of the soil. The low pressure slightly decreased during the 14 days sampling period. A high bubble point of the suction plates of at least 1,000 hPa shortened the period when no water could be collected due to air suction under dry soil conditions. However, in the clay-rich Mehrstedt soil shrinking of the soil induced extensive cracks during summer. Samples could only be collected after moistening and swelling of the clay minerals by precipitation and a regeneration of a coherent soil structure during autumn. Thus, free percolating soil water was collected for a 2.5 years period (winter 2004 to summer 2006).

Spectroscopic properties and radiocarbon analysis

DOC was characterised by its specific UV absorption (SUVA) to assess the degree of complexity and aromaticity of the molecules (Chin et al. 1994; Kalbitz et al. 1999). The UV absorption at $\lambda = 280$ nm was determined with samples adjusted to DOC concentrations of 5 mg L^{-1} (BIO-TEC Instruments, further details in Kalbitz et al. 2003) and SUVA was calculated as $\text{SUVA} = \frac{UV_{280\text{nm}, 5\text{mg}\cdot\text{L}^{-1}}}{5(\text{mg}\cdot\text{L}^{-1})}$ ($\text{L mg}^{-1} \text{m}^{-1}$).

Radiocarbon (^{14}C) concentrations of selected DOC and bulk soil samples were determined with accelerator mass spectroscopy, 3MV AMS (Steinhof et al. 2004) (High Voltage Engineering Europe). DOC samples were selected with three replicates from different suction plates in the non disturbed grassland

stripes at 10 and 110 cm depth for the period 22nd February to 28th April 2005. Some soil water samples from the Mehrstedt site were mixed in order to gain a sufficient amount of C (1 mg). If necessary, samples were decalcified with HCl and freeze dried. Bulk soil was sampled in the non disturbed grassland stripes with a power driven corer with 87 mm diameter in the vicinity of the soil water sampling plots (<30 m distance) in 10, 55 (only at Mehrstedt) and 110 cm depth between November 2003 and November 2004 ($n = 2-3$) (further details on sampling method in Don et al. 2007). The CO_2 evolved during dry combustion of 60–200 mg sample material was reduced to graphite by heating a mixture of H_2 and CO_2 with iron powder at 650°C. The graphite was pressed into targets and measured with AMS. All values were corrected for fractionation of the carbon isotopes during processing using $\delta^{13}\text{C}$ values. The average error was 0.25% modern carbon (pMC). With a mixing model the proportion of recently assimilated C in subsoil DOC was calculated. We assumed no fractionation of ^{14}C during mineralisation or sorption. Since subsoil DOC could derive also from any other parts of the profile aside from the subsoil (110 cm depth) or from recently assimilated C, we could only estimate the maximum proportion of recently assimilated C ($\text{max DOC}_{\text{recent}}$) in subsoil DOC.

$$\text{max DOC}_{\text{recent}} = \frac{^{14}\text{C}[\text{DOC}_{\text{subsoil}}] - ^{14}\text{C}[\text{Bulksoil}_{\text{subsoil}}]}{^{14}\text{C}[\text{DOC}_{\text{recent}}] - ^{14}\text{C}[\text{Bulksoil}_{\text{subsoil}}]} \cdot 100\%$$

The ^{14}C concentrations of recent DOC ($^{14}\text{C}[\text{DOC}_{\text{recent}}]$) were derived from mean atmospheric ^{14}C concentration of a 5 years period prior sampling (107.1 pMC) (Levin et al. 2008).

Meteorological measurements

Meteorological parameters were recorded at a climate station at the Mehrstedt site, including precipitation (heated tipping bucket rain gauge, Thies, Germany). Precipitation for the Kaltenborn site was measured at a nearby weather station (2.5 km away in Bad Salzungen, German Weather Service). Soil moisture was measured in 10 and 65 cm depth in undisturbed soil with permanently installed TDR systems (ML-2x, DeltaT, UK) at both sites, in Kaltenborn only for

5 months. Soil temperature was determined at the Mehrstedt site in 10 and 65 cm depth (PT100, Jumo, Germany).

Water balance and flux model

The water fluxes of the two sites were estimated using a simple water balance model (Hörmann et al. 2007). Since there was hardly any organic layer, a one-compartment storage model was used. The seepage water flux WF was calculated for each sampling period t based on a non-linear model which accounts for water fluxes when soil moisture \leq field capacity (Glugla 1969).

$$WF_t = ((P_t - ET_t + storage_{t-1}) - PWP)^2 \times (c/L^2)$$

P = precipitation during time period t , ET = evapotranspiration during time period t derived from eddy covariance measurements, $Storage$ = changes in soil moisture content between time period t and $t-1$, PWP = permanent wilting point of soil depending on soil texture, c = soil-dependent empirical coefficient after (Glugla 1969), L = soil depth (mean rooting depth, 30 cm).

At the Mehrstedt site, WF was further constrained by soil water content assuming no water flux during summer period when subsoil water content was constantly low. The coefficient c was estimated by inverse modelling using soil moisture content data from both sites for validation. Evapotranspiration ET was determined with a closed path eddy covariance system on a 3 m meteorological tower at the Mehrstedt site (for further details see Don 2007). The relative difference in the amount of collected soil water was used for validation and to check whether ET of the Mehrstedt site can be applied to the Kaltenborn site.

The DOC export (leaching) for i time periods t (2 weeks) was calculated as

$$\sum_{t=1}^i DOC_flux_t = WF_t \times DOC_conc_t$$

to derive annual DOC fluxes. DOC data which were influenced by the installation procedure (settling period) were excluded for the calculations of annual DOC export.

Results

Effects of meteorological conditions on DOC

At Kaltenborn, DOC concentrations near the soil surface (10 cm depth) followed seasonal trends with highest concentrations (max 185 mg L⁻¹) in summer and autumn (Fig. 1). Seasonal DOC dynamics followed soil temperature; i.e. increasing DOC concentrations were measured with increasing soil temperatures (Figs. 1, 2). Trends were less clear at the clay-rich Mehrstedt site with highest DOC concentrations measured in November and December. In Mehrstedt, dry summer conditions prevented any detectable seepage water flux for about 5 months. The first seepage flux after the summer and in Mehrstedt also during the first weeks after installation of the suction plates contained the highest DOC concentrations (Figs. 3, 4). DOC concentration of seepage water leaving the soil system (B–C border) also showed clear seasonal trends but with lower amplitude. Minimum DOC concentrations of the subsoil were found in March (Kaltenborn) and May (Mehrstedt) (Fig. 1).

Both investigated soil types reacted differently to precipitation events as illustrated in Fig. 5: at the Mehrstedt site, precipitation only directly influenced the surface soil moisture content. Subsoil moisture was relatively constant with high values during winter and spring and low values during summer and autumn season. At the Arenosol at Kaltenborn, precipitation directly increased soil moisture in the surface soil and the subsoil (65 cm depth), indicating a rapid seepage water flow through the whole soil profile.

Soil properties and the soil profile

Under similar land use and climatic conditions DOC dynamics depended on soil type with 4–5 times higher DOC concentrations at the Arenosol (Kaltenborn, 14–90 mg L⁻¹) compared to the Vertisol (Mehrstedt, 2–25 mg L⁻¹) (Fig. 6). Mean DOC concentrations decreased with increasing soil depth by 66% (Mehrstedt, 10.9–3.8 mg L⁻¹) and 59% (Kaltenborn, 47.1–19.9 mg L⁻¹) from 10 cm depth to the B–C border (65/110 cm depth). Thus, more DOC was mineralised or retained during transport flux into the subsoil in the Arenosol compared to Vertisol.

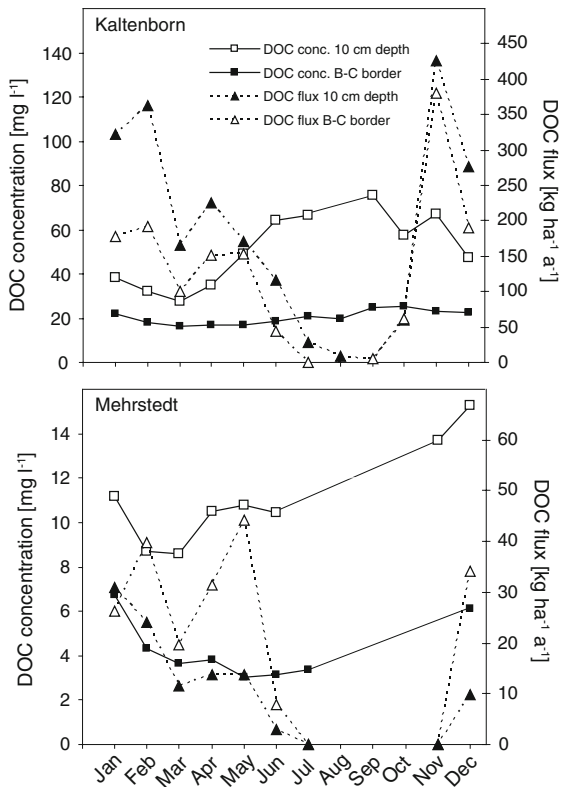
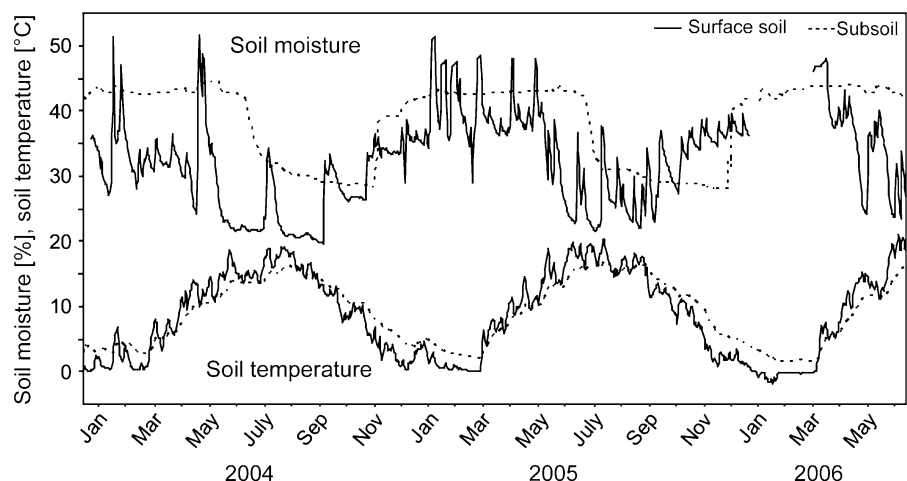


Fig. 1 Seasonal dynamic (monthly means) of DOC concentrations (*quadrante symbols*) and DOC fluxes (*triangle symbols*) in 10 cm depth and at the B–C border in 65/100 cm depth at Mehrstedt and Kaltenborn (*different scales*)

Along with concentration changes in the soil profile DOC chemistry changed as indicated by SUVA values and ^{14}C concentrations (Fig. 7). Surface soil DOC contained higher proportions of aromatic and complex molecules indicated by high

Fig. 2 Seasonal dynamic of soil moisture (%) and soil temperature ($^{\circ}\text{C}$) of the surface soil (10 cm depth) and the subsoil (65 cm depth) at the Mehrstedt site



values of SUVA compared to the subsoil of both sites. At the Mehrstedt site SUVA decreased from 0.022 to 0.014 (10 and 65 cm depth) and from 0.022 to 0.018 (10 and 110 cm depth). Similar trends were found at Kaltenborn but with higher SUVA values throughout the whole profile compared to Mehrstedt. There was hardly any seasonal variability of SUVA at both sites.

Radiocarbon concentrations of DOC also decreased with increasing soil depth from 98.8 to 92.4 pMC (Kaltenborn, 10 and 110 cm depth, respectively) and from 109.5 to 105.9 pMC (Mehrstedt, 10 and 110 cm depth, respectively) (Fig. 7). However, there was a remarkable difference between ^{14}C concentrations of DOC and of SOC from the same soil depth for surface soil and subsoil: higher ^{14}C concentrations of DOC indicate a younger mean age compared to SOC. In particular in the subsoil, the age difference between DOC and SOC was notably high with a mean age difference of more than 4,000 years in Mehrstedt and more than 9,000 years in Kaltenborn. With a mixing model the maximum proportion of recently assimilated C in the subsoil DOC was estimated as 60% for Mehrstedt and 97% for Kaltenborn. Thus, DOC in the subsoil cannot entirely be derived from fresh C sources flushed down the soil profile but at least 40% (Mehrstedt) and 3% (Kaltenborn) have been derived from SOC along the soil profile.

Influence of soil disturbance on DOC

Contrary to expectations, DOC concentrations were 21% (Mehrstedt, $P < 0.001$) and 14% (Kaltenborn, $P = 0.01$) lower at the surface soil of the disturbed

Fig. 3 DOC concentrations (means \pm standard error) in 10 cm depth and at the B–C border (65/110 cm depth) at the Mehrstedt site

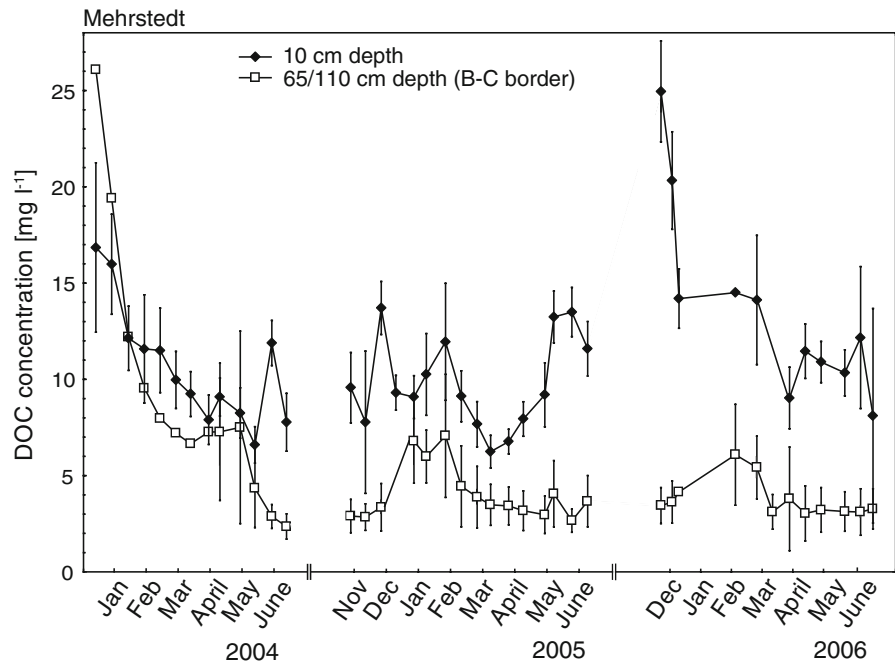
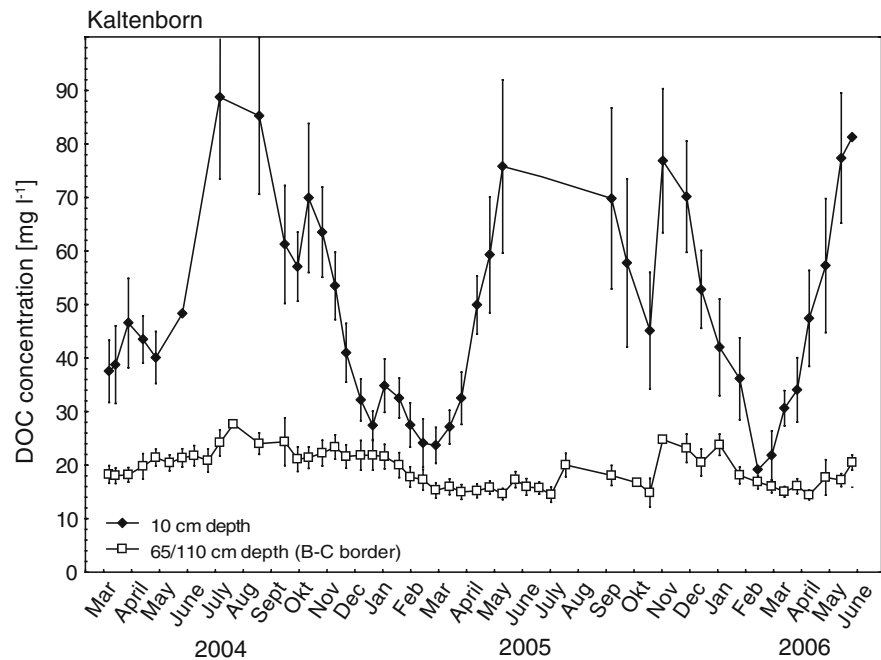


Fig. 4 DOC concentrations (means \pm standard error) in 10 cm depth and at the B–C border (65/110 cm depth) at the Kaltenborn site



ploughing rows than at the neighbouring undisturbed grass stripes (Fig. 6). This significant difference persisted also throughout the second and third year of measurement. Similarly, SUVA indicates a lower aromaticity of DOC in disturbed areas with

significantly lower values at the Kaltenborn in 10 cm depth (mean SUVA of 5.6 in disturbed vs. 6.2 in undisturbed areas, $P = 0.023$). In Mehrstedt there was the same tendency (non-significant) of lower SUVA values in the disturbed ploughing areas.

Fig. 5 Precipitation effects on soil moisture in 10 and 65 cm depth at the Arenosol (Kaltenborn) and the Vertisol (Mehrstedt) for a 5 month period

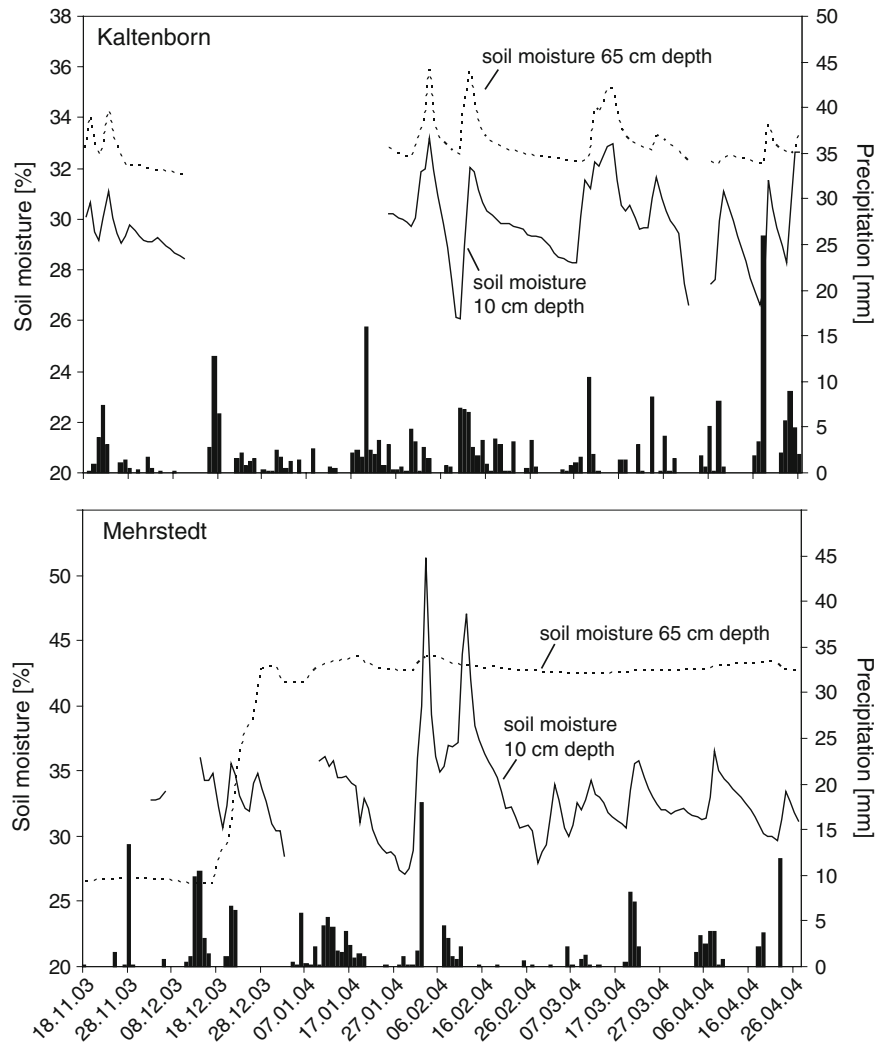
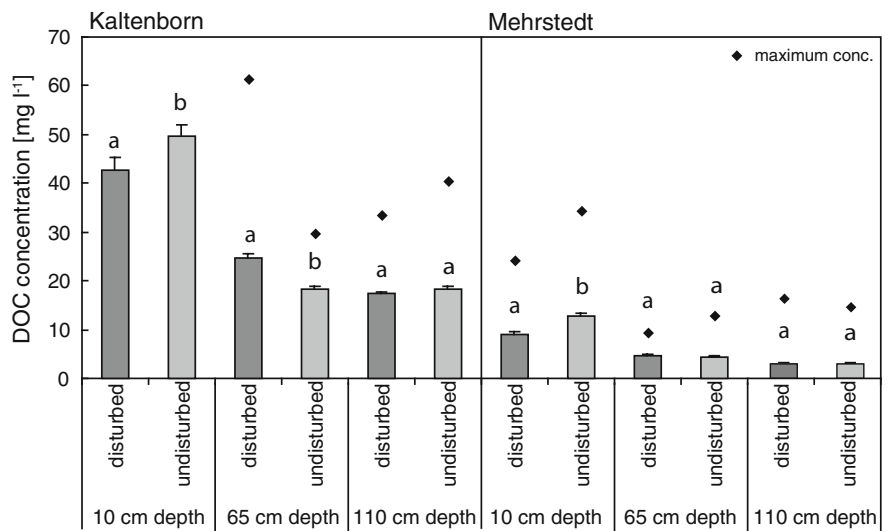


Fig. 6 DOC concentrations (mean \pm standard error and maximum) in disturbed rows (ploughed) and undisturbed grass stripes in 10, 65 and 110 cm depth. Different letters indicate significant differences between disturbed and undisturbed sites



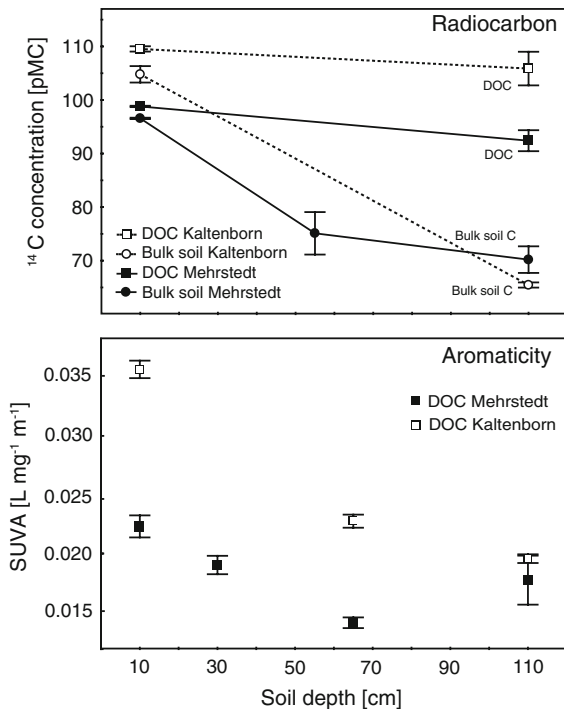


Fig. 7 ^{14}C concentrations (radiocarbon) (means \pm standard error) for DOC and bulk soil and SUVA (means \pm standard error) as measure for DOC aromaticity of DOC and SOC in different soil depths at Mehrstedt (closed symbols) and Kaltenborn (open symbols)

No differences between disturbed and undisturbed areas were found in subsoil DOC samples (65/110 cm depth). Only subsoil DOC concentrations at the Kaltenborn site were higher in the disturbed areas, especially during summer season ($P = 0.003$).

The installation of the suction plates caused a disturbance at aggregate scale which was more intensive than the ploughing of the planting rows. Soil aggregates were crushed and mixed to a suspension that served as contact material between suction plates and soil (see “Materials and methods”). DOC concentrations during the first 3 months after installation were the highest ever measured at the Mehrstedt site (Fig. 3). In contrast, no increased DOC concentrations were detected after plate installation in the sandy soil at Kaltenborn (Fig. 4).

Seepage water and DOC fluxes

A mean annual DOC export at the B–C border of $8 \text{ kg ha}^{-1} \text{ a}^{-1}$ was estimated for Mehrstedt and

$55 \text{ kg ha}^{-1} \text{ a}^{-1}$ for Kaltenborn (Table 1). DOC fluxes in 10 cm depth were more than twice as high with $19 \text{ kg ha}^{-1} \text{ a}^{-1}$ in Mehrstedt and $129 \text{ kg ha}^{-1} \text{ a}^{-1}$ in Kaltenborn. Higher DOC export in Kaltenborn was in line with 64% higher seepage water fluxes as compared to the Mehrstedt site (Table 1). The seasonal dynamic of DOC export was highly dependent on the water fluxes and less dependent on the variability of the DOC concentrations (Figs. 3, 4 and 8). A 75% of the variability of the DOC flux derived from the seasonal variability in the seepage water fluxes and only 25% from variations in DOC concentrations. DOC export and seepage water flux were highly correlated (Pearson correlation coefficient $r^2 = 0.91$ at Kaltenborn and $r^2 = 0.42$ at Mehrstedt) but only weakly with DOC concentrations ($r^2 = 0.01$ and $r^2 = 0.26$). DOC export followed an inverse seasonal trend compared to DOC concentration dynamics: Highest DOC export was determined for winter and early spring season when DOC concentrations were at its minimum but water fluxes were high.

Discussion

Meteorological factors influencing DOC

Seasonality of meteorological factors affected DOC concentrations in all soil depths with maximum concentrations determined during summer and autumn (Kalbitz et al. 2000). At the Mehrstedt site seasonal DOC dynamic was less related to temperature but seems to be obscured by soil properties impact: The clay-rich soil dampened the DOC dynamics due to the adsorption capacity of the clay minerals. Thus, in the subsoil below 65 cm depth minimum DOC concentrations were reached in May and maximum concentrations in December. Even high temperature and plant and microbial activity in June and July with increased DOC at the surface had no direct impact on subsoil DOC (Fig. 1). High DOC concentrations after dry periods can be explained by accumulation of microbial products during dry season that are flushed away with the first seepage water flux in autumn and disruptions to the soil structure (reviewed by Kalbitz et al. 2000). High DOC concentrations after dry periods were found particularly in Mehrstedt where the highly aggregated soil may be perturbed by shrinking and swelling of clay minerals during summer and autumn (Fig. 3).

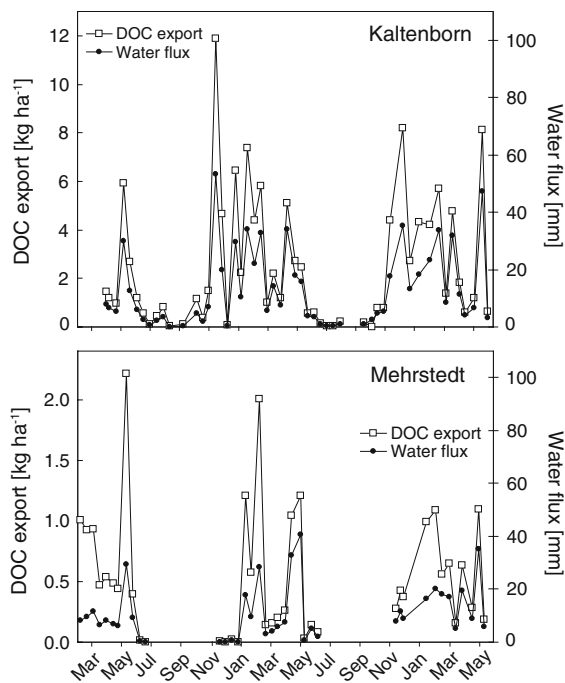


Fig. 8 DOC export (*open symbols*) and water fluxes (*closed symbols*) at the B–C border (65/and 110 cm depth) at Kaltenborn and Mehrstedt (different scales)

The DOC production cannot directly be estimated from our measurements. However, DOC fluxes may indicate DOC productions seasonality, since they represent the fraction of produced DOC which is neither adsorbed nor mineralised. The seasonality of DOC fluxes with highest values during winter and spring was almost complementary to the seasonality of the DOC concentrations (Fig. 1). On one hand, water fluxes control the accumulation of DOC with highest DOC concentrations during periods with low water fluxes (summer season). On the other hand, microbial activity promotes DOC production (Guggenberger and Zech 1993) with enhanced DOC production during summer season as long as the soil water content is no limiting factor. However, microorganisms not only produce DOC but at the same time also consume DOC. We conclude that the seasonal dynamic of DOC fluxes is a result of four processes: DOC production (solubilisation facilitated by microorganisms), DOC consumption (mineralisation by microorganisms), DOC dilution (by precipitation) and DOC transport (seepage water flux), which can not be fully separated from each other in this observational field study.

Soil disturbance and DOC leaching

Disturbance to the soil were found to be important factors for C mineralisation and DOC production (e.g. Hobbie and Likens 1973; Vanveen and Kuikman 1990). Enhanced DOC leaching was found under chisel-ploughed croplands with a C loss of $502 \text{ kg ha}^{-1} \text{ a}^{-1}$ compared to $119 \text{ kg ha}^{-1} \text{ 0a}^{-1}$ under non-disrupted prairie (Brye et al. 2001). A singular disturbance event with deep spate moulder cutting for site preparation of the afforestation did not mobilise significant amounts of DOC from aggregates and did not enhance DOC export of the soil system in our study (Fig 6). Quite the contrary, DOC concentrations were decreased in the disturbed surface soils by 28% (Mehrstedt) and 14% (Kaltenborn). These findings may be explained by decreased substrate availability for DOC production: Soil C profiles of both grassland sites were characterised by a steeply decreasing SOC concentration within the upper 30 cm (Don et al. 2007). Firstly, ploughing decreased surface soil SOC concentrations by uplifting (mixing) of C-poor horizons. Decreased SUVA values in planting rows indicate a shifted DOC source: SUVA was similar to what was found in subsoil DOC samples. Secondly, above-ground biomass was reduced in the disturbed soils by 44 and 22% as compared to the undisturbed grass stripes during summer of the first and second year at the Mehrstedt site (Don 2007) which may have directly influenced surface soil DOC since plant roots and litter provide an extensive source for DOC leaching (Don and Kalbitz 2005; Hagedorn and Machwitz 2007). Lower SUVA values in the disturbed sites compared to the undisturbed sites may also indicate less plant-derived but more SOC-derived DOC. Additionally, secondary effects may control DOC: (1) readily available DOC from fresh litter may control DOC production by enhancing decomposition of old C via priming effect (Fontaine et al. 2007), (2) environmental conditions in the disturbed sites changed due to missing plant cover. Surface soil moisture content on disturbed rows decreased during summer 2004 from 18.6 to 11.1% in Mehrstedt (A. Don, unpublished data) which suppresses microbial activity and DOC production (Thomsen et al. 1999). Thus, disturbance with deep ploughing was not intense enough to make significant amounts of SOC leachable but indirect factors such as soil moisture

and litter production more likely explain lower surface soil DOC concentrations.

Disruption of aggregates was shown to strongly affect DOC production: During the first months after suction plate installation, destruction of soil aggregates generated exceptionally high DOC concentrations in Mehrstedt (Fig. 3). Aggregates in clay-rich soils contain high proportions of SOC which are hardly accessible for microbial attack or leaching unless aggregates are disrupted (Golchin et al. 1994). No effect of soil disruption during suction plate installation was found in Kaltenborn with a single-grain structured sandy soil.

DOC export and translocation through the soil profile

DOC fluxes largely depend on the measurement depth since DOC concentrations are continuously decreasing from the organic layer to the subsoil (e.g. Qualls et al. 2000). To quantify the DOC export of an ecosystem a below ground system border has to be defined, such as the border between the subsoil horizon and the bedrock as it is used in our study. However, all possible systems borders are to some extent arbitrary since DOC fluxes are a continuum from the surface soil to the groundwater ending up in the rivers and oceans. Controls on DOC quality and quantity changes along this pathway have to be better understood. Our study suggest that the high clay content of 35% at the surface soil and up to 70% in the subsoil of the Vertisol (Mehrstedt) promoted DOC adsorption and thus DOC retention and mineralisation (Kalbitz et al. 2000). DOC retention in the soil profile seems to be mostly abiotic sorption and depends on the specific mineral surface area, thus, soil texture and mineralogy (McDowell and Wood 1984; Yano et al. 2005). Additionally, DOC sorption on mineral surfaces is kinetically limited, leading to lower sorption rates under high pore water velocity (Weigand and Totsche 1998). High soil water velocity is typical for sandy soils and has been shown by instantly increasing subsoil water content in the Arenosol (Kaltenborn) after rain events but not in the Vertisol (Mehrstedt, Fig. 5).

The contact time between soil and soil solution is decisive for DOC concentrations since DOC retention in the soil is the precondition for its microbial degradation (McDowell and Wood 1984; Michalzik

and Matzner 1999). Microbial production of extracellular enzymes is an energy demanding investment to facilitate the breakdown of complex, large molecules. It only pays off for individual microbial colonies if substrate is not washed away but residence time is long enough (Ekschmitt et al. 2005). Thus, the retention capacity of clay promotes both DOC mineralisation and DOC stabilisation by adsorption. Higher soil C stocks and younger soil ^{14}C ages at the Mehrstedt site indicate that a higher proportion of assimilated C can be retained and stabilized in the soil. However, retention of DOC can only be maintained permanently if equal amounts of DOC are exported or mineralised or, to some extent, new mineral surfaces are formed by weathering to increase the adsorption capacity of soils (Guggenberger and Kaiser 2003).

Decreasing DOC concentrations with increasing soil depth went along with DOC quality changes such as decreasing aromaticity (SUVA) and increasing mean C ages (^{14}C concentrations) (Fig. 7). DOC quality may be changed due to preferential adsorption and mineralisation of specific DOC fractions (Kaiser et al. 2004). In contrast, recent studies suggest that DOC is derived from SOC by “autarkic” DOC production in each soil horizon with little direct DOC translocation through the soil profile (Fröberg et al. 2007; Steinbeiss et al. 2008). Changes in DOC quality with soil depth may reflect SOC quality changes throughout the soil profile. ^{14}C concentrations indicate that at least 40% of subsoil DOC at the Mehrstedt site has to derive from SOC along the soil profile and not from recently assimilated C from the surface soil. High ^{14}C concentrations of DOC and a maximum proportion of 97% recently assimilated C in subsoil DOC in Kaltenborn indicate a stronger connections between surface soil and subsoil in the sandy soil.

DOC export cannot be estimated from SOC stocks assuming high DOC export in SOC rich soils. SOC stocks at the Mehrstedt site (89 t C ha^{-1} 0–60 cm depth) were almost twice as high as in Kaltenborn (46 t C ha^{-1}) but DOC export in Mehrstedt was 85% smaller than in Kaltenborn (8 and $55 \text{ kg C ha}^{-1} \text{ a}^{-1}$, respectively). Thus, annual DOC export in Mehrstedt equals 0.01% of the SOC stocks and 0.12% in Kaltenborn. Other studies found DOC export being largest from C rich organic soil and organic layers (forest floor) (Hope et al. 1994; Michalzik and Matzner 1999). High DOC exports in forests and

peatlands derived from non-physically stabilised C sources which may explain high export rates and direct correlations between C stocks and DOC export. In mineral soils, like in most temperate grassland soils, major parts of SOC are not accessible for DOC leaching. Large differences in ^{14}C concentrations between SOC and DOC from the same horizon, in particular in the subsoil (Fig. 7), suggest that most SOC is spatially separated from soil water fluxes, e.g. within fine pores or aggregates, and cannot be leached as DOC. Since dissolution is the precondition for microbial decomposition, these SOC fractions may be more stabilised.

The Vertisol at the Mehrstedt site is characterised by a pronounced soil shrinking during summer when no low-pressure could be maintained in the suction plate-system. Even a high bubble point of the suction plates could not prevent a gap of sampling of several months without collectable soil water. A hysteresis effect during rewetting of the soil resulted in prolonged periods without collectable soil water during autumn. Thus, annual DOC fluxes could be underestimated due to missing soil water fluxes during summer and early autumn. However, a highly negative water balance during summer periods indicates that there was no water fluxes at the B–C border. Soil water measurements in 65 cm depth showed that rain events during summer did not influence these soil depths (Fig. 2). Thus, DOC flushing with summer storm events is unlikely to generate DOC fluxes at deeper soil depth.

A positive correlation between DOC and water flux has been found in many temperate and boreal catchments (Harrison et al. 2008; Hope et al. 1994; Mertens et al. 2007). DOC fluxes are controlled by hydrological conditions on plot scale (Kalbitz et al. 2000) and on watershed scale (Eimers et al. 2008) especially in water limited grassland ecosystems like in our study. Seven times higher DOC export at Kaltenborn compared to Mehrstedt may partly be explained by 64% higher seepage water fluxes and 20% higher precipitation in Kaltenborn. DOC loss from the soil system followed the high discontinuity of the water fluxes with a high correlation between water flux and DOC export. Thus, the uncertainty of the climatic water balance of about 10–30% is directly carried forward to the DOC export estimations.

DOC export was only $8 \text{ kg ha}^{-1} \text{ a}^{-1}$ at Mehrstedt and $55 \text{ kg ha}^{-1} \text{ a}^{-1}$ at Kaltenborn which is about 1%

(Mehrstedt, Vertisol) and 8% (Kaltenborn, Arenosol) of mean net ecosystem productivity (Don 2007; Soussana et al. 2007). Differences between both sites due to the soil type and texture are in line with reported 8–9 times higher DOC exports from a Orthic Haplohumod with coarse sandy texture compared to a Typic Hapludult with a sandy loamy texture from cultivated grass-clover swards (Vinther et al. 2006). A high correlation between DOC export and soil CN was found for biome scales including semi-terrestrial soils like peatlands (Aitkenhead and McDowell 2000). Only small differences in soil CN between both sites of our study suggest that CN is no adequate predictor for DOC fluxes in our terrestrial soils. Soil pH controls the charge density of dissolved organic molecules and the complexation with polyvalent cations such as iron and aluminium, with decreased solubility of DOC at lower pH like at the Kalteborn site (Kalbitz et al. 2000). Higher SUVA values at the Kaltenborn site suggest more complex molecule structures than at the Mehrstedt site which would decrease solubility and degradability (Don and Kalbitz 2005). In fact, these parameters hardly explain the large differences in DOC concentrations and fluxes between both sites; in contrast, higher DOC would be expected in Mehrstedt than in Kaltenborn. It is not possible to draw general conclusions from these two case studies but it seems that texture as a physico-chemical soil parameter dominates the influence of other parameters on DOC export at our two sites.

DOC export is a missing C flux in many ecosystem C balance studies. Net ecosystem productivity derived from land surface-atmosphere exchange measurements (eddy covariance method) would be overestimated without accounting for DOC losses. Among the rare permanent grassland DOC studies, winter season DOC export of $42\text{--}118 \text{ kg C ha}^{-1}$ was determined for a grassland on clay-rich Gleysols (McTiernan et al. 2001). This export was higher than in our study but may be explained by higher seepage water fluxes (362 mm) compared to water fluxes in our study (177 and 290 mm). Limitations for decomposition due to water logging may increase DOC content in Gleysols but were rarely found on our two sites. Catchment-based estimates of DOC export from temperate grasslands range from 1.6 to $89.8 \text{ kg C ha}^{-1} \text{ a}^{-1}$ with high values explained by high peatland coverage and low values derived from catchments with low runoff (<60 mm) (Hope et al.

1994). DOC export was considered a missing C flux for the European C balance (Siemens 2003). In fact, DOC export from agricultural land has been rather vague and more effort is needed to get estimates for different land use, climate regimes and particular for different soil types.

Conclusions

- The dominant factor for DOC export is the seepage water flux. Seasonal variability and total DOC export amounts were directly related to the soil water fluxes.
- Single events of soil disturbance by ploughing did not increase DOC export on a time scale of several years. However, aggregate disruptions lead to exceptionally high DOC release after the installation of the suction plates.
- Soil physicochemical parameters such as texture determined DOC retention in our soils and DOC concentration profiles. We propose to further elucidate the potential of soil texture as a predictor to upscale DOC fluxes for terrestrial soils excluding histosols.
- Our data suggest that DOC leaching and transport is largely decoupled from SOC and cannot be derived from SOC stock data. For the investigated two terrestrial soils DOC export was seven times higher in the SOC poor site than in the site with high SOC stocks.
- High differences in ^{14}C concentrations between bulk soil and DOC indicate that DOC production, transport and adsorption are spatially limited to aggregate interfaces and preferential flow paths with little connection and exchange with SOC outside these “hot spots”.

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